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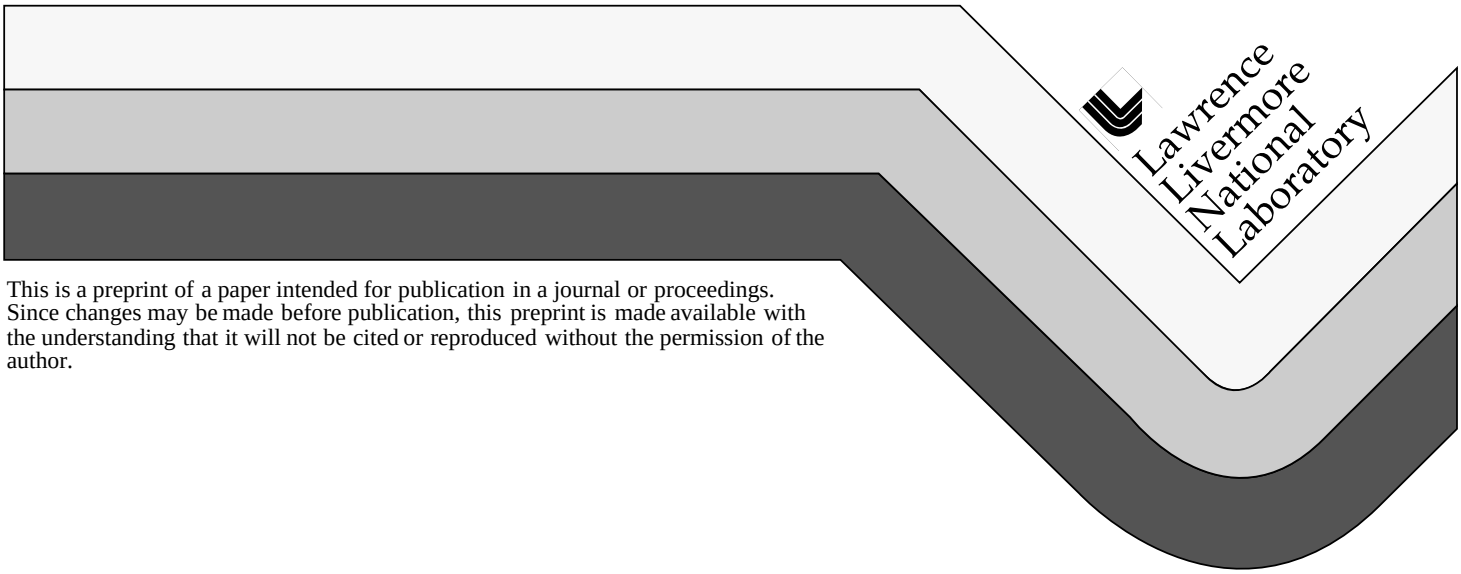

Applications and Development of High Pressure PEM Systems

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Abstract

Many portable fuel cell applications require high pressure hydrogen, oxygen, or both. High pressure PEM systems that were originally designed and developed primarily for aerospace applications are being redesigned for use in portable applications. Historically, applications can be broken into weight sensitive and weight insensitive cell stack designs. Variants of the weight sensitive designs have been considered to refill oxygen bottles for space suits, to provide oxygen for space shuttle, to provide oxygen and/or reboost propellants to the space station, and to recharge oxygen bottles for commercial aviation. A long operating history has been generated for weight insensitive designs that serve as oxygen generators for submarines. Exciting future vehicle concepts and portable applications are enabled by carefully designing lightweight stacks which do not require additional pressure containment. These include high altitude long endurance solar rechargeable aircraft and airships, water refuelable spacecraft, and a variety of field portable systems. High pressure electrolyzers can refill compressed hydrogen storage tanks for fuel cell powered vehicles or portable fuel cells. Hamilton Standard has demonstrated many high pressure PEM water electrolyzer designs for a variety of applications. Electrolyzers with operational pressures up to 3000 psi (20.7 MPa) are currently used for US Navy submarine oxygen generators. An aerospace version has been demonstrated in the Integrated Propulsion Test Article (IPTA) program. Electrolyzers with operational pressures up to 6000 psi (41.4 MPa) have also been demonstrated in the High Pressure Oxygen Recharge System (HPORS). Onboard oxygen generator systems (OBOGS) that generate up to 2000 psi (13.8 MPa) oxygen and refill breathable oxygen tanks for commercial aviation have been designed and successfully demonstrated. Other hardware applications that require high pressure PEM devices are related to these proven applications.

High Pressure PEM Product Background

PEM fuel cells were the first fuel cells used in space (1 kW unit for Gemini), and have accumulated over 5000 stack hours in space. Other PEM fuel cells developed for aerospace applications include a 350 W unit for a biosatellite spacecraft, a 5 kW prototype for a NASA technology program, and a 3 kW unit for the US Navy aerostat. Space fuel cell applications generally require both hydrogen and oxygen storage, airborne applications can consider use of atmospheric oxygen, while undersea applications (also of aerospace PEM technology) must concentrate on supplying the oxygen.

PEM electrolyzers are used in the SSN-21 (Seawolf class submarines) at 3000 psi (20.7 MPa) for life support oxygen. This high pressure oxygen generation technology is enabling wherever oxygen may be scarce: under water, in mining, at high altitude, etc. Over 18 million cell hours have been accumulated in the Royal Nuclear Navy without a single malfunction. PEM electrolyzers are being developed for space station metabolic oxygen. This line of development traces directly back to Gemini hardware, which was developed by General Electric (GE), and continues to date through Hamilton Standard (HS), Space and Sea Systems, a division of United Technologies Corporation.

PEM electrolysis produces both hydrogen and oxygen, in the exact stoichiometric proportion that fuel cells consume. Many PEM cell designs require pumps, but the simplest system variants can eliminate all moving parts (except the poppets inside valves and water expulsion containers) by suitable modifications to the electrolysis cell itself. Electrolyzers that do not require a high pressure or circulating pump, known as “static feed” electrolyzers, have been developed for use with propellant generators for small satellites since the mid-1970’s. Up to 1000 psi (6.9 MPa) hydrogen and oxygen gas generation pressures have been demonstrated using high pressure water. HS has since improved this capability with proprietary cell designs that can produce high gas pressures from low pressure water.

LLNL adopted a reversible aerospace PEM technology, available only from HS before 1998, in order to solve the very challenging problem of propelling a solar power aircraft through the night. The extreme weight-sensitivity of this advanced vehicle application favors the combination of electrolysis and fuel cell modes of operation in the same PEM cell design. Energy storage applications typify many portable applications of fuel cells, and reversibility allows portable fuel cells to provide their own fuel (dispensing with the requirement for a hydrogen infrastructure). PEM static feed reversible (unitized) fuel cells (URFCs) were demonstrated (over 700 cycles, 2.1 MPa) for small satellite energy storage (1-4). More recent testing at Lawrence Livermore National Laboratory (LLNL) proved high cycle life (>2000 cycles) and high performance URFCs (2-5).

Static Feed PEM Electrolyzer and Reversible Cell Design

Static water feed electrolysis is an innovation that can generate high pressure gases without pumps. This form of electrolysis transports water (by osmosis and diffusion) between the water chamber and the oxygen electrode, where electrolysis occurs. The generated hydrogen gas diffuses into the water chamber and must be prevented from masking the water feed barrier from the water supply. Two approaches for preventing water feed barrier masking have been demonstrated. One approach maintains the water pressure above hydrogen pressure, preventing hydrogen gas from coming out of solution. The other approach uses an electrochemical hydrogen pump that returns hydrogen to the hydrogen cavity to prevent its accumulation on the water side, and thereby allows operation with a low pressure water feed (4,6). The diffusion of hydrogen and oxygen gas through the electrolysis cell is a loss mechanism, but does not form explosive gas mixtures since the diffusing gases react on the same catalyst which split protons from water. Reaction heat is conducted away from the cell to an external heat sink by a metal foil. Hamilton Standard static feed cells use edge current collection and can be stacked in electrical series by external connections.

Advanced High Pressure PEM Systems

The development of the hydrogen-oxygen-water electrochemical system is sufficiently perfected technologically to contribute to the advancement of many aerospace goals. HS proprietary PEM cell designs (known as SPE®) are used in water electrolyzers that are in production for submarine oxygen generation, commercial hydrogen generation, and Space Station life support. The last ten years of progress have shown significant advances in electrolyzer hardware development, mostly with respect to inherent maximum cell stack operating pressure, unaided by pressure domes or other auxiliary equipment. Today's technology makes possible electrolyzer assemblies that directly generate the gases at up to tank storage pressure (tested to 12 MPa for terrestrial applications) with only the additional stack weight required to contain the pressure. There is an overall weight savings at the system level when compared with earlier designs, due to the elimination of a number of support components. This new electrolyzer hardware capability is very attractive for small craft propulsion when it is deemed practical to produce gases in-situ. For spacecraft applications, water is easy to store and launch, while the specific impulse of hydrogen-oxygen thrusters is superior to all other nontoxic propellant combinations. Producing propellant as needed also reduces the storage volume required, a significant bonus of this high-pressure technology (4,5,7-12).

Water Supply in High Pressure Systems

Today's high pressure electrolysis technology was made possible by the early development of perfluorosulfonic acid, ion exchange membrane (PEM) which displays superior mechanical and chemical properties compared to its predecessors, sulfonated polystyrene-divinylbenzene membranes. The electrode reactions for water electrolysis are those typical for an acid electrolyte, where protons are the main charge carrier. The membrane is, macroscopically, an impermeable member in the assembly. This situation inside an electrochemical cell is quite different from the porous diaphragm separators of industrial electrolyzers in that it creates two distinct cavities in the assembly where the process water can be supplied.

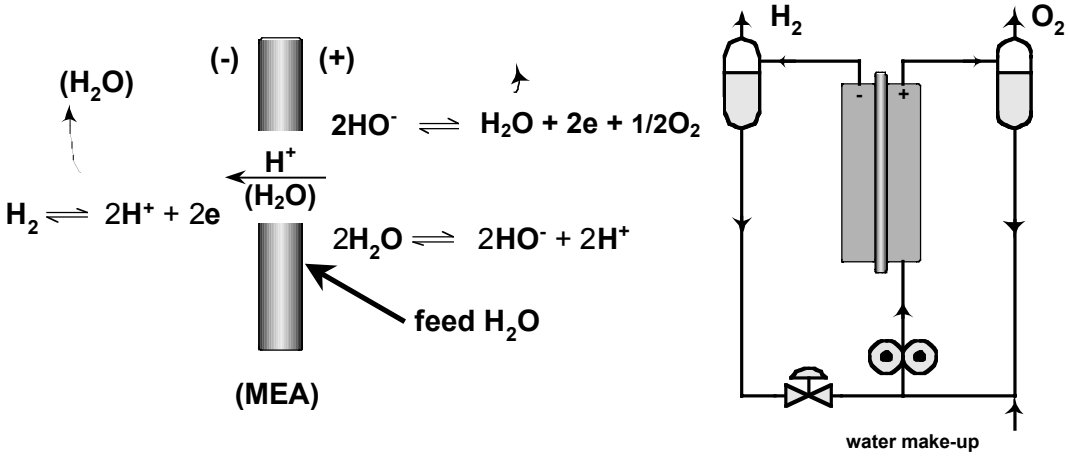


Figure 1. Schematic of Anode Feed Electrolysis

When the water is supplied at the anode side (oxygen side), it dissociates so that protons migrate to the cathode under the effect of the electric field, and hydroxyl ions are discharged to form neutral oxygen gas. The protons are also discharged at the cathode forming neutral hydrogen gas. Feed water is usually circulated through the anode cavity in order to perform the thermal management for the electrolysis stack at the same time it provides the process water. Oxygen gas has to be separated from this feed water before it can be used or stored. The proton migrating through Nafion® type PEM membranes is hydrated to an extent depending on the membrane water content. As a result, the hydrogen generated in an anode feed electrolysis cell contains liquid water that also (like the oxygen in anode feed) needs to be separated before either gas can be used or stored. A simplified schematic is shown in Figure 1. The proton migration through the membrane in the anode feed mode creates a net water flux from anode to cathode that needs to be counteracted. The schematic example shown in Figure 1 requires two phase separators, while the cathode water is returned to the

anode stream through a controlling valve. Because the water concentration gradient across the membrane is very small, membrane ohmic resistance is not excessively affected by the current density. The proton flux actually ensures that water is continuously passed through the membrane, thus maintaining the membrane water content. Consequently, anode fed electrolysis systems are capable of the highest current densities attainable with PEM technology (in excess of 3000 A/ft^2 [3.2 A/cm^2]), and of the lowest cell potentials for a given operating temperature and pressure.

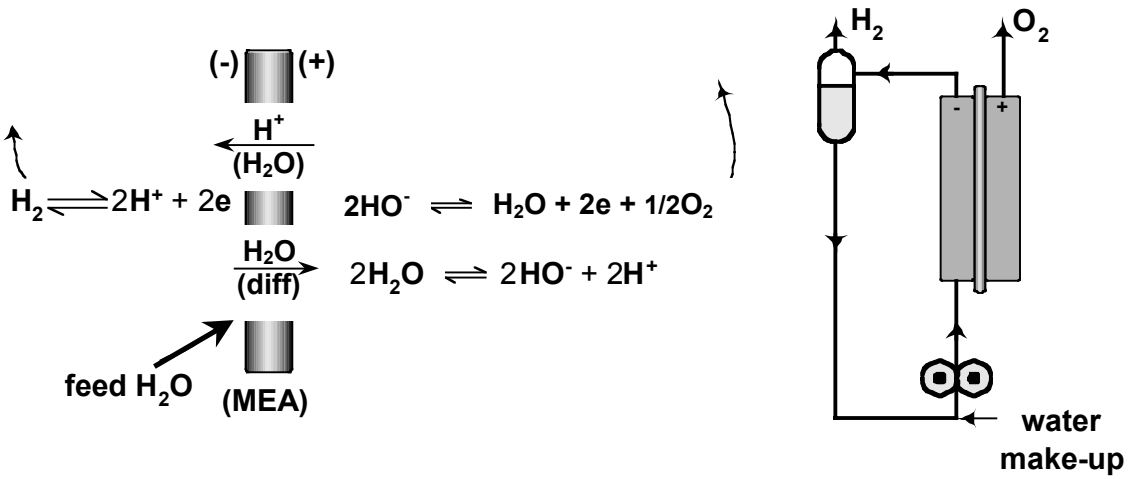


Figure 2. Schematic of Cathode Feed Electrolysis

PEM electrolysis systems can be simplified somewhat if the process water is fed on the cathode side, as shown in Figure 2. The reactions are the same but water reaches the anode side by diffusion. The proton is still hydrated to enable its migration through the membrane, thus returning some of the water to the cathode side. While the diffusion flux is controlled mainly by the concentration gradient across the electrolysis membrane-electrode assembly (MEA), the cell current (or current density) controls the electro-osmotic flux associated with the proton migration. As these two processes oppose each other, a limiting condition occurs which balances the water returned with protons almost exactly with the water diffusing to the anode.

Because the effective diffusion of feed water from cathode to the anode electrode requires a concentration difference, cells operated in cathode feed mode typically develop a large water gradient across the membrane, increasing the cell resistance and, consequently, the cell potential. Cell performance in cathode feed reflects this water transport as a limit in the cell's current density capability, beyond which a small increment in the current density causes an accelerated increase in the cell potential. Such runaway increases in cell potential threaten the very integrity of the cell assembly. The advantage of operating an electrolysis

system in cathode feed mode, as shown in Figure 2, is that the oxygen stream is free of liquid water.

Phase separation is generally not a major system design consideration unless the equipment has to operate in a zero-G environment or unless the additional weight of ancillary components (especially heavy to withstand high pressures) severely limits the applicability of the technology. Cathode feed designs eliminate one phase separator at the expense of the maximum operating current density. To eliminate the second phase separator, a built-in water metering system must be designed that will deliver water only to the extent that it is being consumed. At the same time liquid water must be restrained so that it is not entrained with the product gases. In the electrolyzers tested using solid polymer electrolyte technology, water is contained behind a semi-permeable membrane, a water feed barrier (WFB), that allows the water to reach the electrolysis membrane only to the extent that the water already present there has been consumed.

The water in a WFB design, known as a "static feed" design, migrates under a chemical potential gradient between the barrier membrane surface facing the cell cathode and the cathode surface of the electrolysis membrane. In general, the reactions in a WFB cell are the same, as shown in Figure 3, but the overall electrolysis system is greatly simplified. Both the hydrogen and oxygen gases are rather dry if electrolysis is conducted at low enough temperature. Otherwise, the product gases are saturated at the process temperature, in equilibrium with the water vapor pressure above the electrolysis membrane. The same current density issues apply as for the cathode feed option. However, the need to have additional gradients established across the cell to effect the water transport across two membranes further adds to the penalty one pays in performance for the benefit of a very simple system.

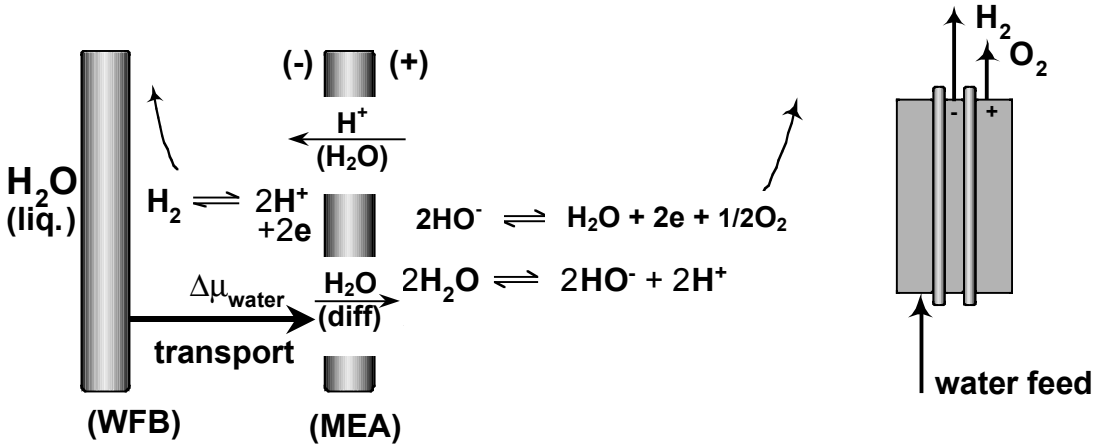


Figure 3. Schematic of Static Feed Electrolysis

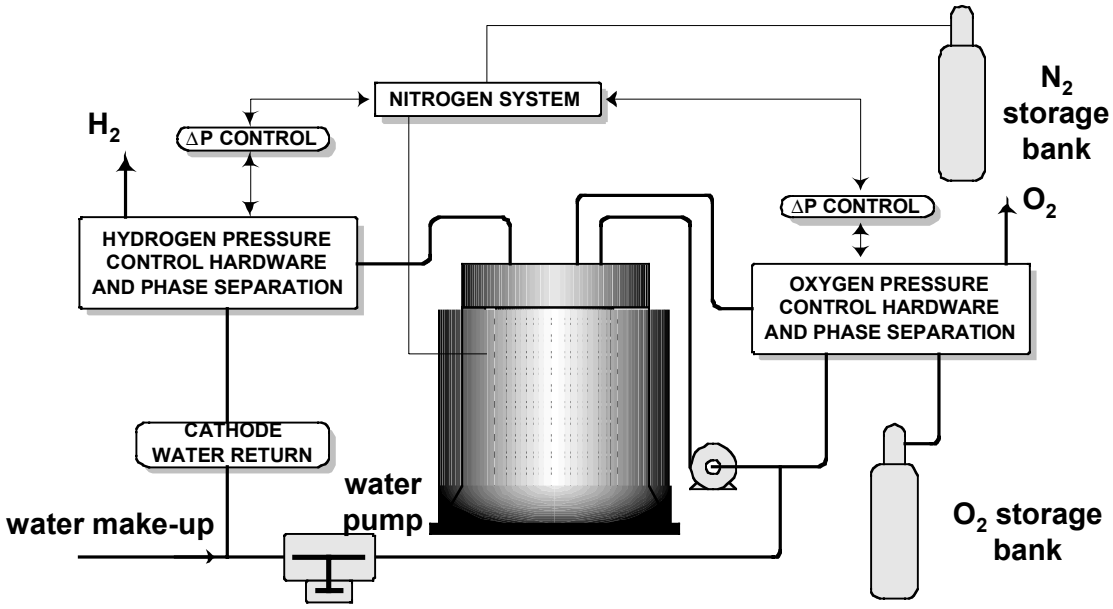
The first generation of high-pressure passive feed water electrolyzers were described earlier (6) and their electrical performance reported. However, they

were heavy assemblies, hardly suitable for flight-testing. A new generation of passive water feed electrolysis stacks is emerging, lighter, better electrical efficiency and more durable, for flight applications up to 130 atm (13 MPa).

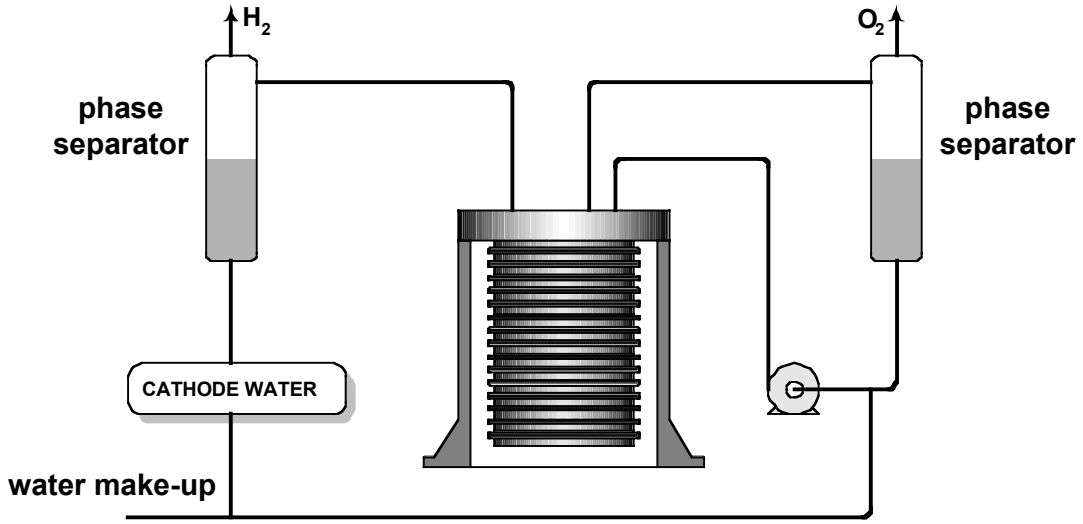
High Pressure Systems Options

The gases produced through water electrolysis are very pure and should not require additional processing. It is advantageous therefore to match electrolysis pressure with that required for further processing or storage. The initial high pressure solution adopted for the SPE® electrolysis stacks was to enclose the entire assembly in a pressure vessel with a blanket of inert gas for pressure management. That solution was derived from earlier electrolyzer and fuel cell systems using liquid electrolyte held in a porous matrix. This approach assigns the job of providing for both electrolysis stack and system safety to the system designer. Systems employing this option were consequently heavy and needed constant attention, in addition to requiring supplemental resources in the form of high pressure nitrogen gas. The Oxygen Generating Plant (OGP) developed for the U.S. Navy used this approach. Subsequent work on oxygen generators was directed towards improving the pressure capabilities of the electrolysis stack itself. The potential for system simplification through a more capable stack assembly is summarized in Figure 4, (13-16), which uses a generic life support application as an illustration. Improved overboard pressure capability allows the elimination of the pressure vessel. If that were the only improvement, however, the nitrogen system would still be needed to maintain the balance between the anode and cathode pressure cavities unless the membrane support on either side was capable of withstanding a differential pressure equal to the full system pressure.

A low pressure stack installed in a high pressure system must be operated in a balanced pressure mode and requires nitrogen to be used by the control system if the system operating pressure exceeds the mechanical capability of the electrolysis membrane support. In addition to this nitrogen, differential pressure limits require that both gases be generated at about the same pressure even if only one is actually being stored. For the life support application illustrated in Figure 4, the membrane support capability was improved to allow the oxygen to be produced at near-ambient pressure while the hydrogen remains at the elevated pressure. A high pressure feed pump is eliminated and the anode circulating loop is greatly simplified. The only function not provided for in these re-designed stacks is high pressure gas storage. With these improvements, the HS electrolysis system has evolved into (trans)portable hardware that can be used in far more applications than originally envisioned, although these applications all use the same hydrogen/oxygen/water system for energy conversion.



Low Pressure Stack in a High Pressure System



High Pressure Stack in a High Pressure System

Figure 4. Schematic of Advanced Generic Life Support Application

Capabilities built into the electrolyzer hardware have a direct impact on the system configuration and operating reliability. The more capable the stack, the simpler and more reliable the system can be. Where system weight is a design factor, the simplified system offers a head start on achieving the weight goals. Although the increased capability of the electrolysis stack design does not improve the process efficiency, it impacts the system in a way by making the overall application easier to manage.

Rechargeable power sources using the hydrogen-oxygen-water electrochemical cycle have been on the wish list of aerospace system designers for decades. System analyses always foretell energy density advantages over secondary batteries whenever capacities exceed a nominal threshold value, provided the storage pressure can be made high enough. A few attempts made using electrochemical cells with alkaline electrolyte failed to demonstrate a practical longevity. Although Hamilton Standard is not pursuing reversible electrolysis cell hardware at this time, HS design studies show that high pressure technology is the ideal hardware vehicle for such an endeavor. While a reversible stack has to be sized for the required power output, the overall system energy density is determined mainly by the maximum pressure capability of reactant gas storage. For weight sensitive applications, the ability to generate the gases at the required storage pressure is quite advantageous because it eliminates the mechanical compressor, and electrochemical compression is more energy efficient than adiabatic compression, resulting in a modest but meaningful power saving. Because no mechanical/moving parts are involved, properly designed high pressure electrochemical hardware has a much longer maintenance-free life than any mechanical device.

LLNL Design Study

In 1998, a single 0.23 ft^2 static water feed electrolyzer that was built by Hamilton Standard was demonstrated at LLNL. Its single cell uses an electrochemical hydrogen pump which allows hydrogen pressure to exceed water pressure. The unit was manufactured in the mid-1980's and remained in storage for a dozen years before being activated in 1998. The 1998 performance duplicated performance obtained during the mid-1980's. Several units containing up to seven cells were tested during the 1980's, accumulating many thousands of hours of experience. This demonstrator is rated at 200 psi hydrogen and oxygen gas generation pressure, using ambient water pressure. The same basic design with reenforced frames was recently demonstrated successfully at 1000 psi at NASA Marshall Space Flight Center.

A trade study was performed by Hamilton Standard in support of LLNL spacecraft applications. Its focus was electrolysis propulsion for spacecraft (known as a "Water Rocket"), and its deliverables included three preliminary designs of lightweight, high pressure PEM electrolyzers. This small spacecraft propulsion application requires an electrolyzer that would be suitable for charging almost all portable, hydrogen-fueled or oxygen-constrained applications. Static Feed PEM electrolyzers with nominal electrical power inputs of 50, 100, and 200 W, were sized in sufficient detail to predict all component masses. Prior technology that could address the same applications includes vapor feed and high pressure anode feed electrolyzers. In 1986 a vapor feed electrolyzer was

assembled with polysulfone frames and a series of electrical and thermal conducting rings. More recently, anode feed electrolyzers have been tested that provide higher pressure capability than the polysulfone frame design at lower weight per cell. These two types of proven electrolyzers were incorporated into a cell design that supplies high pressure oxygen and hydrogen free of liquid water. This has been accomplished using the Hamilton Standard static feed electrolysis cell configuration and high pressure hardware design.

Requirements for LLNL's cell stack design included production of hydrogen and oxygen at 2000 psi (13.8 MPa), with water supplied at ambient pressure, long operating life, and as lightweight as practical. Three separate cells were designed for three different power ratings, with active areas of 22.2 in^2 (143 cm^2), 44.5 in^2 (287 cm^2), and 88.9 in^2 (574 cm^2). The 50 W, 100 W, and 200 W units utilize the same 16 cell arrangement, so that the voltage drop for each stack was held constant across all three designs. Each cell is equipped with thermal management provisions in the form of heat conduction tabs to carry heat out of the stack, and power tabs for electrical communication between individual cells, and with the power source. Individual cells are electrically isolated within the stack and connected externally in series. Stack weight in the 100 W unit was minimized with a combination of 16 cells each with a 0.019 ft^2 (17.7 cm^2) active area. For this configuration, the cell outer diameter is 2.63 inches and end dome flange diameter is 3.57 inches.

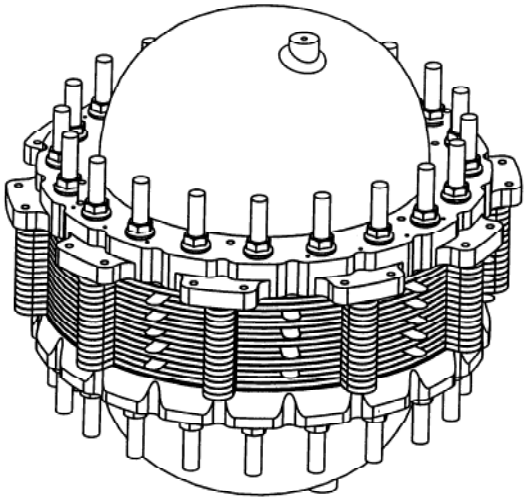


Figure 5. LLNL Design for 200 Watt Electrolyzer

The 200 W cell stack design is shown in Figure 5. Heat transfer sheets are composed of thin metal foils. Individual cells have a cavity which is ported to the end dome to maintain uniform compression and contact throughout the stack. This cavity removes the need for separate pressure pads, which helps reduce the weight of the cell stack. Stainless steel tie rods and lock nuts are preloaded to compress the seal area and contain the high pressures in the oxygen and

hydrogen compartments. The three different sized units use 8, 12, and 20 tie rods respectively.

End dome assemblies are made of high strength-to-weight metal, and each includes a bladder with a metal pressure plate to transfer compressive forces to the active areas, maintaining uniform electrical contact throughout. The upper end domes are ported to allow for the inlet and outlet gases, which eliminates the need for a central fluid plate. This design saves on both the overall weight and size of the cell. The end dome operating pressure is 3000 psi (20.7 MPa).

Concerns for the cells designed in this paper study include maintaining sufficient electrical contact through the active area of each cell. Since the membrane material tends to expand through the cell active area, as the seal area on the membrane is compressed, it is difficult to estimate active area gaps and compression requirements prior to assembling hardware. Another concern is the material selection for the end dome bladders. Although the material selection will not cause a considerable variance of the weight of the cell, the material performance during the life of the cell is critical. Other issues that await resolution after the assembly of a test unit include the amount of creep in the tie rods and membrane degradation.

Table 1: Conceptual Design of 2000 psi Static Water Feed Electrolyzer

Input Power	50 Watt	100 Watt	200 Watt
Number of Cells	16	16	16
Active Area/cell [in^2 (cm^2)]	1.39 (8.97)	2. 78 (17.9)	5.56 (35.9)
Mass of stack [lb (kg)]	1.15 (0.523)	1.74 (0.789)	2.83 (1.28)
Oxygen pressure [psi (MPa)]	2000 (13.8)	2000 (13.8)	2000 (13.8)
Hydrogen pressure [psi (MPa)]	1980 (13.7)	1980 (13.7)	1980 (13.7)
Water pressure [psia (MPa)]	15 (0.10)	15 (0.10)	15 (0.10)
Operating Temp. [°F (°C)]	70-160 (21-71)	70-160 (21-71)	70-160 (21-71)
Net electrolysis at 120°F (49°C)	2.8 g H_2O/hr	5.6 g H_2O/hr	11.1 g H_2O/hr

Detailed drawings and models for the manufacture of cell parts for the 200 Watt system have been generated for the 2000 psi static water feed electrolyzer design. These drawings and models include: upper compression dome, lower compression dome, water screen frame, hydrogen screen frame, oxygen screen frame, membrane & electrode, water feed barrier, separator frame, and insulator. Water is supplied from a surface tension tank or bladder tank at low pressure. (Space applications demand component technologies that operate in zero gravity, which imposes no further requirements on static feed cell designs but does demand special water tanks.) No rotating equipment, no moving parts except valve poppets (and water expulsion device motion) will be required to assist with the water supply for static feed cell designs. Heat is transported by conduction to a heatsink. The number of valves is minimized: just two valves,

each one controlling one gas as it is transferred to a fuel cell (or rocket engine). Two electrolyzer gas outlet valves operate only on startup and when safing the system. Gas dryers are optional and may be employed to prevent ice formation inside equipment. (This consideration applies to many field-portable and outdoor applications. Note that only ~0.2 lbm of water is delivered with the gases for each 100 lbm of water electrolyzed at 120 °F. Dryers for 0.2 lbm of water can weigh ~3 lbm, so they may not be mass efficient. Freeze prevention by strategically placed heaters may be more mass effective. Lightweight regenerative dryers have been investigated by Hamilton Standard that employ PEM membranes and high pressure technology.

The 200 W static feed electrolyzer design can be modified for electric energy storage, by making it into a reversible cell. The modifications include providing a temporary water storage volume into each cell to accomodate the product water produced during discharge periods. A mass increase of only ~25% was estimated to be required in order to achieve reversible operation. The crucial modification requires no additional mass: changing the cell oxygen electrode catalyst to one that is tailored for cell reversibility.

Aerospace development is likely to implement and prove the performance of lightweight, high performance PEM stacks (both electrolyzers and reversible cells) similar to those just described. Figures 6-11 document six already developed aerospace PEM stacks and systems. Progress anticipated over the next year or two is likely to provide the ideal gas generators and reversible PEM stacks to enable high pressure energy storage and portable fuel cell applications.

Illustrations of High Pressure PEM Systems

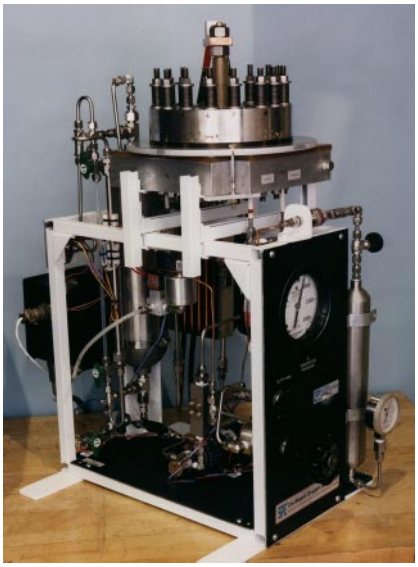


Figure 6. Demonstration hardware for the On-Board Oxygen Generating System (OBOGS). One cell using the reinforced polysulfone hardware, capable of 3000 psi proof. The cathode feed system was designed to be very portable; it required only water and a power source at the site.

Figure 7. OBOGS demonstration system package. The system could fill a 250 ml tank to 1800 psi or vent directly to ambient through a back pressure regulator. It was capable of automatic start and stop based on a pre-determined number of conditions. Sized for 200 ASF (0.22 A/cm^2) operation.

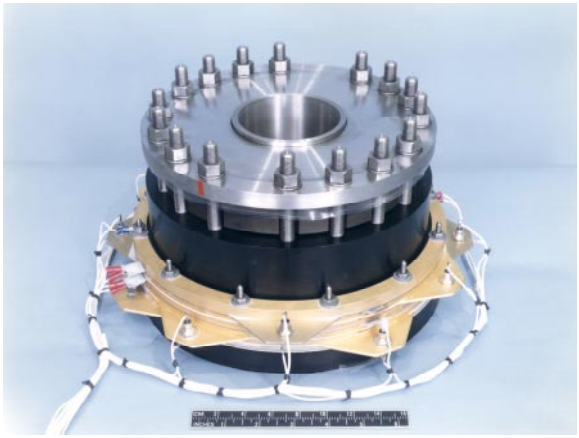


Figure 8. Static (passive) water feed 5-cell assembly. Capable of 1000 psi, balanced pressure operation. Maximum current density tested for was about 300 ASF at 120°F. It used the old fashioned, polysulfone hardware (0.23 ft^2 active area) with external reinforcements instead of a dome. It is the full size assembly of the single cell that was tested at LLNL

Figure 9. Integrated Propulsion Test Article (IPTA). Built as a demonstrator for a propulsion electrolyzer. Anode feed, 3000 psi balanced pressure. Used the old style hardware and therefore required the dome and the nitrogen system (not shown). It was in all respects a scaled down version of the oxygen generator for the submarine.

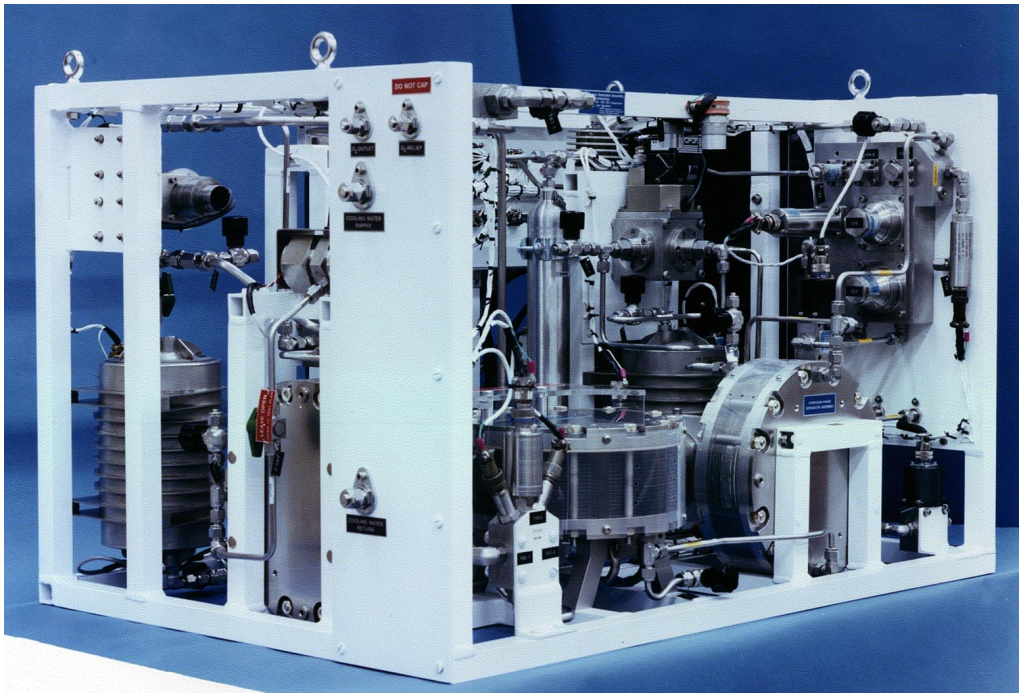


Figure 10. Oxygen Generating Assembly (OGA) for NASA, second iteration. Anode feed. The design included passive, integrated membranes, zero-G phase separators for both anode and cathode sides. The system is very portable; it contains its own system controller in the same package. Requires only water and power from the site. Capable of continuous or cyclic mode operation to simulate the space station rotation. Designed to generate hydrogen at 40 psi and oxygen at near ambient pressure. The operating current density for the “light” side of the cycle is 200 ASF. During the dark side it maintains a 1 A trickle current

Figure 11. High pressure, anode feed hardware. Metal laminate assembly capable of 3000 ASF current density. Developed for life support, the technology is conducive to extremely lightweight developments. The assembly was designed for anode feed operation with the hydrogen being produced at high pressure and the oxygen at ambient. The water make-up does not require a high pressure pump. The same technology is used for the 200 W passive water feed electrolyzer for LLNL. Portable lightweight electrolyzers are taken one step further towards new applications.

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